22P016-O

Observation of laser ablation phenomena in super critical water

超臨界水中におけるレーザーアブレーション現象の観測

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1. Introduction

Conventional laser ablation, which is used for thin film deposition and nanoparticles syntheses, has been carried out in gases. Recently, laser ablation in liquids attracts significant attention of many researchers as a new technique to produce nanoparticles. In this work, we employed supercritical water as an alternative medium for laser ablation. Supercritical water has high chemical reactivity and properties of gas and liquid. Thus the supercritical water is attractive state as a new laser ablation medium. In this work, we observed the optical emission intensity from laser ablation plasmas produced in supercritical water and a bubble-like phenomenon which was observed after the disappearance of the laser ablation plasma. We compared the experimental results in supercritical water with those in liquid water and gas.

2. Experimental

The experimental apparatus is schematically shown in Fig. 1. A cell which was compatible with a high pressure of 35 MPa and a high temperature of 500 ^{o}C was filled with distilled water. YAG laser pulses at a wavelength of 1064 nm irradiated a Ti target installed in water from the normal direction. The duration of the YAG laser pulse was 10 ns. The YAG laser beam was focused using a lens. When we observed optical emission from ablation plasma in liquid water at a pressure of 30MPa and a temperature of 230^{o}C, the laser energy was adjusted at 36 mJ/pulse, while in the case of supercritical water at a pressure of 30 MPa and a temperature of 430^{o}C, an laser energy of 360 mJ/pulse was employed. The difference in the laser energy was to compensate the different sizes in the ablation craters formed in water and supercritical water. The different sizes of the ablation craters were due to the fact that the tight focusing of the laser beam in the vertical direction was impossible in supercritical water. When we observed bubble-like phenomena in the supercritical water by shadowgraph imaging,

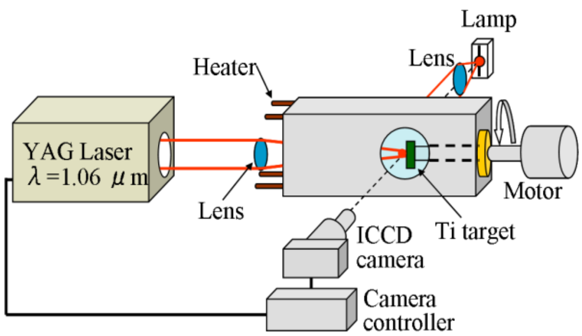

Fig 1.Diagram of experimental setup

a laser energy of 22.4 mJ/pulse was employed. The optical emission image of the ablation plasma was captured using a charge-coupled device camera with a gated image intensifier (ICCD camera) from the side of the high-pressure, high-temperature cell. The gate width was 2ns. In shadowgraph imaging, the ablation space was illuminated by flash lamp light, and the transmitted flash lamp light was captured using the same ICCD camera with a gate width of 5 ns. The temporal variations were obtained by changing the delay time between the irradiation of the YAG laser pulse and the trigger to the gate of the ICCD camera.

3.Results and discussion

Figure 2(a) shows normalized temporal variations of the optical emission intensities from laser ablation plasmas produced in liquid water (30 MPa, 230^{o}C) and supercritical water (30 MPa, 430^{o}C). The optical emission intensities plotted in Fig. 2 were obtained by integrating the optical emission images spatially. As shown in the figure, the rise parts of the optical emission intensities were rather similar in liquid water and supercritical water, while the temporal decay of the optical emission intensity in supercritical liquid water was much slower than that in liquid water. Figure 2(b) shows spatial distributions of the optical emission

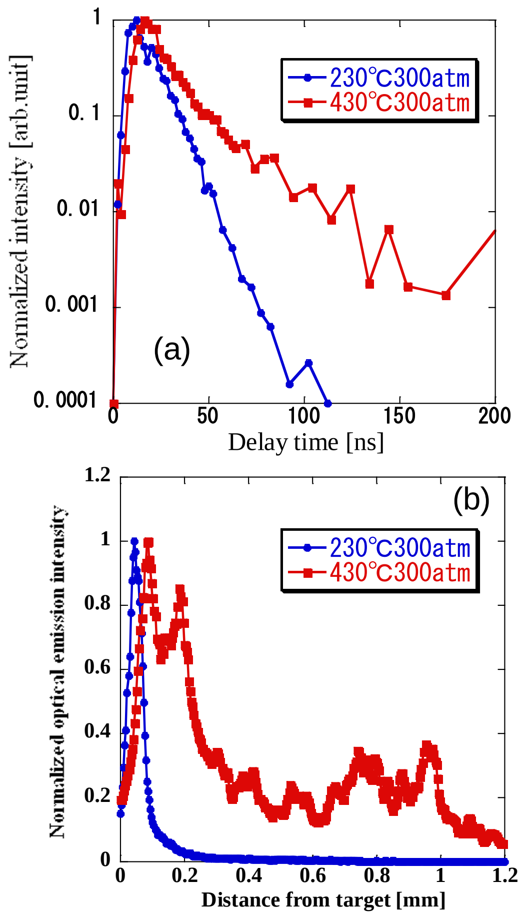

Fig. 2 (a) Temporal variations and (b) spatial distributions of optical emission intensities from laser ablation plasmas produced in liquid water (30 MPa, 230℃) and supercritical water (30 MPa, 430℃).

intensities. The delay times for capturing the optical emission images were 14 and 16 ns in liquid water and supercritical water, respectively. These delay times corresponded to the maximum optical emission intensities. As shown in the figure, the optical emission region in supercritical water was much broader than that in liquid water. The experimental results shown in Fig. 2 indicate less tighter confinement and less significant quenching in supercritical water than liquid water. These characteristics are reasonable since supercritical state has smaller density and is classified into the intermediate between liquid and gas.

In liquid-phase laser ablation, we observe the formation of a cavitation bubble after the disappearance of the plasma with optical emission. In the case of laser ablation in supercritical water, we also observed a bubble-like phenomenon as shown in Fig. 3. We detected the formation of a bubble-like density hollow at 4 μs after laser

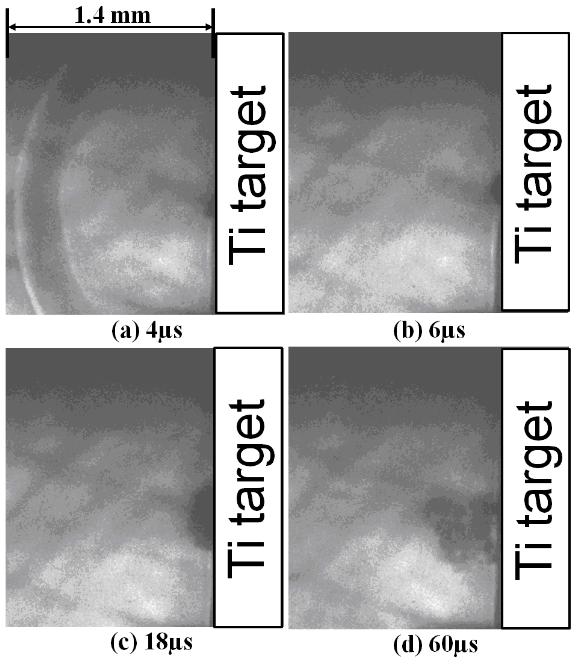

Fig. 3 Shadow graph images of the ablation space in supercritical water (30 MPa and 430℃) at various delay times after laser ablation..

ablation. The density hollow expanded with time, and the maximum size was observed at 18 μs. Remarkable differences from the cavitation bubble observed in liquid-phase laser ablation are summarized as follows; 1) the maximum size of the bubble-like density hollow was larger than that of a cavitation bubble in liquid-phase laser ablation at the same pressure, 2) the bubble-like density hollow expanded for a longer time than a cavitation bubble, and 3) the bubble-like density hollow never shrank after the maximum size while a cavitation bubble shrinks and comes to the collapse. The last shape of the density hollow observed in supercritical water was not the collapse but a kind of extinction into supercritical water. The density hollow in supercritical water is remarkably different from the cavitation bubble, but is rather similar to phenomena observed in the behavior of ambient gas in gas-phase laser ablation [1]. This would be also due to the fact that supercritical state is the intermediate between liquid and gas.

References

[1] K. Sasaki and H. Watarai, Jpn. J. Appl. Phys. **45**, L447 (2006).